

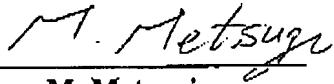
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DECLARATION

I, Makoto Metsugi, c/o MIYAZAKI & METSUGI, of Daido Seimei Building, 5-4, Tanimachi 1-chome, Chuo-ku, Osaka, Japan certify that I am the translator of the documents in the Japanese language, Japanese Patent Application No. 2002-381184, attached hereto and that to the best of my knowledge and belief the following is a true and correct translation thereof.

April 24, 2007

  
M. Metsugi

Filing Date:December 27, 2002  
REFERENCE No.=NPX1020025 Patent Appln.No. 2002-381184 Page: 1/2

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【Inventor】  
【Address or Residence】 c/o SANYO ELECTRIC CO., LTD.  
5-5, Keihanhondori 2-chome,  
Moriguchi-city, Osaka  
【Name】 Masaharu ITAYA  
【Inventor】  
【Address or Residence】 c/o SANYO ELECTRIC CO., LTD.  
5-5, Keihanhondori 2-chome,  
Moriguchi-city, Osaka  
【Name】 Masahide MIYAKE  
【Inventor】  
【Address or Residence】 c/o SANYO ELECTRIC CO., LTD.  
5-5, Keihanhondori 2-chome,  
Moriguchi- city, Osaka  
【Name】 Masahisa FUJIMOTO  
【Applicant】  
【Identification Number】 000001889  
【Name】 SANYO ELECTRIC CO., LTD.  
【Agent】  
【Identification Number】 100095382  
【Patent attorney】  
【Name】 Makoto METSUGI  
【Appointed Agent】  
【Identification Number】 100086597  
【Patent Attorney】  
【Name】 Chikara MIYAZAKI

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**【Official Fee】**

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**【List of Attached Documents】**

【Title of Attached Document】 Specification 1

【Title of Attached Document】 Abstract 1

**【Proof】** Demanded

[DOCUMENT NAME] SPECIFICATION

[TITLE OF THE INVENTION] ELECTROLYTE FOR NONAQUEOUS BATTERY,  
METHOD FOR PRODUCING THE SAME, AND ELECTROLYTIC SOLUTION FOR  
NONAQUEOUS BATTERY

5 [CLAIMS]

[CLAIM 1] An electrolyte for a nonaqueous battery  
consisting essentially of magnesium  
bistrifluoromethanesulfonimide.

[CLAIM 2] A method for producing an electrolyte for a  
10 nonaqueous battery comprising the step of reacting magnesium  
carbonate or magnesium hydroxide with an imide compound to  
produce the electrolyte for a nonaqueous battery.

[CLAIM 3] A method for producing an electrolyte for a  
nonaqueous battery comprising the step of reacting magnesium  
15 carbonate or magnesium hydroxide with  
trifluoromethanesulfonimide to produce magnesium  
bistrifluoromethanesulfonimide.

[CLAIM 4] An electrolytic solution for a nonaqueous battery  
comprising:  
20 magnesium bistrifluoromethanesulfonimide; and  
an organic solvent and/or a room temperature molten salt having  
a melting point of 60°C or less in which the magnesium  
bistrifluoromethanesulfonimide is dissolved.

[CLAIM 5] The electrolytic solution for a nonaqueous  
25 battery according to claim 4, wherein at least one kind selected

from the group consisting of a cyclic carbonate, a chain carbonate, a cyclic ether, a chain ether, a cyclic ester and a chain ester is used as the organic solvent.

[CLAIM 6] The electrolytic solution for a nonaqueous  
5 battery according to claim 4 or 5, wherein an ammonium salt is used as the room temperature molten salt.

[DETAILED EXPLANATION OF THE INVENTION]

[0001]

[TECHNICAL FIELD OF THE INVENTION]

10 The present invention relates to an electrolyte which is useful for a nonaqueous battery such as a magnesium ion battery, a method for producing the electrolyte and an electrolytic solution using the electrolyte.

[0002]

15 [PRIOR ART]

Lithium ion batteries having high energy density have been put to practical use. Attentions have been focused on magnesium and calcium as an active material having high energy density the same as that of lithium.

20 [0003]

However, magnesium salts and calcium salts soluble in an organic solvent are few, and as for the magnesium salts, magnesium organohaloaluminate is only examined (NON-PATENT DOCUMENT 1).

[0004]

25 [NON-PATENT DOCUMENT 1]

Nature, 407, 724 (2000), D. Aurbach, Z. Lu, A. Schechter, Y. Gofer, H. Gizbar, R. Turgeman, T. Cohen, M. Moshkovich and E. Lev.

[0005]

5 [PROBLEMS TO BE SOLVED]

It is an object of the present invention to provide an electrolyte for a nonaqueous battery which is useful for a magnesium ion battery or the like and is a magnesium salt soluble in an organic solvent, and a method for producing the electrolyte.

10 It is further another object of the present invention to provide an electrolytic solution for a nonaqueous battery using the electrolyte.

[0006]

[MEANS FOR SOLVING THE PROBLEMS]

15 An electrolyte for a nonaqueous battery according to the present invention consists essentially of magnesium bistrifluoromethanesulfonimide [ $Mg((CF_3SO_2)_2N)_2$ ].

[0007]

The present inventors found that the magnesium bistrifluoromethanesulfonimide can be dissolved in an organic solvent, and the organic solvent in which the magnesium bistrifluoromethanesulfonimide is dissolved shows sufficient conductivity of about  $10^{-3} S cm^{-1}$  as an electrolytic solution of a battery. The present invention was accomplished based on 25 this finding.

[0008]

The electrolyte according to the present invention can be used for a nonaqueous battery such as a magnesium ion primary battery and a magnesium ion secondary battery.

5 [0009]

An electrolytic solution for a nonaqueous battery according to the present invention includes the magnesium bistrifluoromethanesulfonimide as the electrolyte according to the present invention. Specifically, the magnesium bistrifluoromethanesulfonimide is dissolved in an organic solvent and/or a room temperature molten salt having a melting point of 60°C or less.

[0010]

Examples of organic solvents in which the electrolyte according to the invention can be dissolved include a cyclic carbonate, a chain carbonate, a cyclic ether, a chain ether, a cyclic ester and a chain ester. The organic solvents may individually be used or a mixture of two or more kinds thereof may be used.

20 [0011]

Examples of cyclic carbonates include ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), trifluoropropylene carbonate (TFPC) and fluoroethylene carbonate (FEC). Examples of chain carbonates include dimethyl carbonate (DMC), diethyl carbonate (DEC) and methyl ethyl

carbonate (MEC). Examples of cyclic ethers include sulfolane (SL), tetrahydrofuran (THF) and crown ether (12-crown 4, 15-crown 5, 18-crown 6 or the like). Examples of chain ethers include dimethoxyethane (DME), ethoxymethoxy ethane (EME) and 5 diethoxyethane (DEE). Examples of cyclic esters include  $\gamma$ -butyrolactone ( $\gamma$ -BL), valerolactone (VL) and angelica lactone (AL). Examples of chain esters include methyl formate (MF), methyl acetate (MA) and methyl propionate (MP).

[0012]

10 Examples of room temperature molten salts having a melting point of 60°C or less in which the electrolyte according to the present invention can be dissolved include salts made by combining a cation selected from ammonium, imidazolium, pyrazolium, triazolium, thiazolium, oxazolium, pyridinium, 15 pyridazinium, pyrimidinium and pyrazinium, and an anion selected from  $\text{BR}_4^-$ ,  $\text{PR}_6^-$ ,  $\text{RSO}_3^-$ ,  $(\text{RSO}_2)_2\text{N}^-$  and  $(\text{RSO}_2)_3\text{C}^-$  (wherein R represents a halogen element,  $\text{CF}_3$ ,  $\text{C}_2\text{F}_5$ , or an alkyl group or an aryl group having other electron-attracting groups). Specifically, examples of ammonium salts include trimethylpropyl ammonium-bis-(trifluoro methylsulfonyl) imide (TMPA-TFSI) 20 (( $\text{CH}_3$ )<sub>3</sub> $\text{N}^+(\text{C}_3\text{H}_7)$  ·  $\text{N}^-(\text{SO}_2\text{CF}_3)_2$ ). Examples of imidazolium salts include 1-ethyl-3-methyl imidazolium-2, 2, 2-trifluoro-N-(trifluoro methylsulfonyl) acetamide (( $\text{C}_6\text{H}_{11}\text{N}_2$ )<sup>+</sup> · ( $\text{CF}_3\text{CO}$ ) $\text{N}^-(\text{SO}_2\text{CF}_3)$ ). Examples of pyrazolium salts include 1, 25 2-dimethyl-4-fluoropyrazolium-tetrafluoroborate (( $\text{C}_5\text{H}_8\text{N}_2\text{F}$ )<sup>+</sup> ·

$\text{BF}_4^-$ ). Examples of pyridinium salts include 1-ethyl pyridinium-2, 2, 2-trifluoro-N-(trifluoro methylsulfonyl) acetamide ( $(\text{C}_7\text{H}_{10}\text{ N})^+ \cdot (\text{CF}_3\text{CO})\text{N}^- (\text{SO}_2\text{CF}_3)$ ) .

[0013]

5 The magnesium bistrifluoromethanesulfonimide dissolved in the organic solvent or the room temperature molten salt is not limited to particular amount. The magnesium bistrifluoromethanesulfonimide is dissolved in an amount to cause the conductivity required such as the conductivity of  $10^{-3}$   
10  $\text{S cm}^{-1}$ .

[0014]

A method for producing an electrolyte for a nonaqueous battery according to the present invention comprises the step of reacting magnesium carbonate or magnesium hydroxide with an  
15 imide compound to produce the electrolyte for a nonaqueous battery.

[0015]

When the magnesium bistrifluoromethanesulfonimide which is the electrolyte for a nonaqueous battery according to the  
20 present invention is produced, the magnesium bistrifluoromethanesulfonimide can be produced by reacting magnesium carbonate or magnesium hydroxide with trifluoromethanesulfonimide.

[0016]

25 By using the electrolytic solution for a nonaqueous battery

according to the present invention, a positive electrode made of  $Mg_xMo_3S_4$  and a negative electrode made of Mg, a magnesium ion secondary battery can be composed.

[0017]

5 [EMBODIMENTS OF THE INVENTION]

Hereinbelow, the present invention will be described in detail by way of examples, although the present invention is not limited to the following examples.

[0018]

10 (Example)

Trifluoromethanesulfonimide ( $(CF_3SO_2)_2NH$ : hereinbelow, referred to as "HTFSI") was dissolved in 1 liter of water to prepare a 1 mole / liter (1M) solution. Magnesium carbonate ( $MgCO_3$ ) was added to the solution at 1:2 mole ratio of  $MgCO_3$  15 to HTFSI while the solution was stirred. The magnesium carbonate reacted with the HTFSI as follows to form magnesium bistrifluoromethanesulfonimide, carbon dioxide and water.

[0019]

[Formula 1]

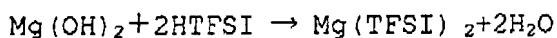


[0020]

When magnesium hydroxide was used in place of the magnesium carbonate, the magnesium hydroxide reacted with the HTFSI as follows to form magnesium bistrifluoromethanesulfonimide and water.

[0021]

[Formula 2]



[0022]

5 After the present inventors confirmed that the magnesium carbonate was entirely dissolved, water and carbon dioxide were removed by depressurization by using a rotary evaporator to obtain white magnesium bistrifluoromethanesulfonimide. The magnesium bistrifluoromethanesulfonimide obtained was  
10 vacuum-dried at 220°C for 8 hours to obtain anhydrous magnesium bistrifluoromethanesulfonimide.

[0023]

The magnesium bistrifluoromethanesulfonimide obtained was added to propylene carbonate (PC), a mixture solvent (EC:DMC) 15 of 1:1 volume ratio of ethylene carbonate (EC) to dimethyl carbonate (DMC),  $\gamma$ -butyrolactone ( $\gamma$ -BL) and butylene carbonate (BC) respectively. The present inventors confirmed that the magnesium bistrifluoromethanesulfonimide is dissolved in the solvents. Additionally, the conductivity of each solution in 20 which 1M (1 mole/liter) of the magnesium bistrifluoromethanesulfonimide was dissolved was measured.

The results were shown in Table 1. The moisture value in 1M of each solution was 100ppm or less.

[0024]

25 When the magnesium bistrifluoromethanesulfonimide was

added to trimethylpropyl ammonium trifluoromethanesulfonimide

(TMPA-TFSI) as a room temperature molten salt, the present inventors confirmed the dissolution of the magnesium bistrifluoromethanesulfonimide. Additionally, the

5 conductivity of 0.5 M (0.5 mole / liter) of the room temperature molten salt solution was measured and the result was shown in Table 1. The conductivity shown in Table 1 was measured at 25°C.

[0025]

[Table 1]

Solvent	Conductivity ( $\times 10^{-3}$ S cm $^{-1}$ )
PC	3.31
EC:DMC	5.83
$\gamma$ -BL	6.87
BC	1.34
TMPA-TFSI	2.50

10

[0026]

As shown in Table 1, the conductivity of each solution was in the range of  $1.34 \times 10^{-3}$  to  $6.87 \times 10^{-3}$  S cm $^{-1}$ . The conductivities were almost equal to that ( $7.90 \times 10^{-3}$  S cm $^{-1}$ ) of a mixture solvent of 1:1 volume ratio of EC to DEC (diethyl carbonate) which was a typical electrolytic solution for a lithium ion battery and in which 1M of LiPF<sub>6</sub> was dissolved. Therefore, the solutions can be used as an electrolytic solution for a nonaqueous battery.

15 20 [0027]

[EFFECT OF THE INVENTION]

The present invention can provide an electrolyte and an electrolytic solution for a nonaqueous battery which are useful for a magnesium ion battery or the like. Additionally, an electrolyte for a nonaqueous battery as a magnesium salt which 5 is soluble in an organic solvent or the like can be produced in a convenient process by the method for producing according to the present invention.

[DOCUMENT NAME] SUMMARY PAPER

[SUMMARY]

[OBJECT] To obtain an electrolyte for a nonaqueous battery which is useful for a magnesium ion battery, a method 5 for producing the electrolyte, and an electrolytic solution for a nonaqueous battery.

[MEANS FOR SOLVING THE PROBLEM] An electrolyte for a nonaqueous battery according to the present invention consists essentially of magnesium bistrifluoromethanesulfonimide. An 10 electrolytic solution for a nonaqueous battery according to the present invention includes the magnesium bistrifluoromethanesulfonimide, and an organic solvent such as a cyclic carbonate, a chain carbonate, a cyclic ether and a chain ether or an ordinary temperature molten salt having a melting 15 point of 60°C or less in which the magnesium bistrifluoromethanesulfonimide is dissolved.

[SELECTED FIGURE] None